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Light scattering by slightly non-spherical particles on surfaces

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We investigate the shape dependence of the scattering by dielectric and metallic particles on surfaces by considering particles whose free-space scattering properties are nearly identical. The scattering by metallic particles is very strongly dependent upon the shape of the particle in the region near where the particle and surface contact. The scattering by dielectric particles displays a weaker, but nonetheless significant, dependence on particle shape. These results have a significant impact on the use of light scattering to size and identify particles on surfaces.

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The inspection of surfaces for particles is an important step in the development of contamination-free manufacturing in the semiconductor, optics, and data storage industries. Tools based upon laser light scattering are often used to detect such contaminants. One of the challenges is to accurately identify the size and material of contaminants that are found, so that their source can be identified and the contamination problem solved. Multiple scattering channels, such as multiple detectors in a single-wavelength scanning system, are employed to provide information that can lead to particle identification and sizing. The development of these systems requires accurate scattering models for ideal and non-ideal particles, and appropriate interpretation of the results from those models.

In this Letter, we present some theoretical results that indicate that for metallic particles on semiconducting surfaces, the scattering by slightly nonspherical particles can deviate substantially from spheres having identical volumes. The scattering turns out to be very dependent upon the shape of the particle near the point where it contacts the surface.

An accurate theory for the scattering of light by spherical particles above a surface was developed by Bobbert and Vlieger[1] in 1986. Their method, which relies upon the calculation of the reflection matrix of the surface in terms of the vector spherical harmonics or spherical Debye potentials, can be straightforwardly extended to axisymmetric[2] and even arbitrary particles,[3] by using the T-matrix approach of Barber and Hill.[4] For this study, we implemented the theory described in Ref. 2, and include the presence of a substrate coating by explicitly incorporating the appropriate reflection coefficients. We include the coating partly because such a layer normally exists on silicon and partly because its presence improves the rate of convergence of the solution. Other methods have also been used to calculate the scattering by non-spherical features on surfaces.[5–8] The T-matrix method used here was compared with the discrete-dipole method[8] using spherical and ellipsoidal (oblate and prolate) particles, yielding identical results within the accuracy of the discrete-dipole approximation.

While the calculations were carried out for a variety of conditions, we will limit our presentation to some specific ones. The wavelength of 488 nm was chosen because the Ar^+ laser line is commonly used in semiconductor wafer inspection tools. We use p-polarized (electric-field in the plane of incidence) light incident at an oblique angle $\theta_i = 60°$, since an electric field perpendicular to the surface most strongly displays the effects that we show, and because oblique incidence and p-polarization yields larger scattering cross sections and are thus used in inspection applications. The particle materials will be limited to polystyrene (index $n = 1.605$) and aluminum (index $n = 0.73+5.93i$), since polystyrene is often used as a standard for calibrating instruments and has optical properties typical of many insulators, and aluminum is a possible contaminant in wafer processing and has optical properties [$\mathrm{Re}(n) \leq 1$] which enhance the effects which we show. We choose silicon with a 1.5 nm native oxide for the substrate, since it is a common material to inspect and, with its high index ($n = 4.37 + 0.08i$), yields a strong particle-substrate interaction. Scattering will be shown only in the plane of incidence, as a function of the scattering angle θ_r, measured from the surface normal; the results in out-of-plane directions are similar but are more difficult to visualize. The sign of θ_r is such that the specular condition occurs when $\theta_r = \theta_i$.

A number of studies have investigated the scattering by ellipsoids[2,3,9,10] and cylinders[10] on surfaces. However, since such particles scatter differently in free space than spheres of equal volume, the observations do not illustrate the effect the surface plays in the scattering. We choose instead to illustrate the effect of shape by two classes of axisymmetric particles, both of which have free space scattering properties very similar to spheres of equal volume. By choosing particles in this manner, we can effectively separate the effects that have to do with the global particle shape from those which result from the interaction with the surface.

Particle shape is parameterized by the function $\tilde{r}(\theta,\phi)$ describing the surface of the particle in spherical coordinates, where the polar angle θ is measured from the particle-to-substrate direction. Since we consider only axisymmetric particles, $\tilde{r}(\theta,\phi) = r(\theta)$. The first class of particles consist of spheres that are dented (truncated) by a small amount d:

$$r_{\mathrm{T}}^{\mathrm{d}}(\theta) = \begin{cases} r_0 & (\theta \geq \theta_0) \\ (r_0 - d)/\cos\theta & (\theta < \theta_0) \end{cases} \qquad (1)$$

where θ_0 is given by $\cos^{-1}[(r_0 - d)/r_0]$. We chose $r_0 = 30$ nm and $d = 1.2$ nm, creating a contact area with a radius 8.4 nm. The truncation can also be aimed away from the surface so that $r_{\mathrm{T}}^{\mathrm{u}}(\theta) = r_{\mathrm{T}}^{\mathrm{d}}(-\theta)$; in this case, there is zero contact area, and the radius of curvature is the same as the sphere radius r_0. The shape of the second class of particle is based upon the Chebyshev polynomials and is given by

$$r_{\mathrm{C}}(\theta) = r_0[1 + \xi\cos(m\theta)], \qquad (2)$$

where r_0, ξ, and m parameterize the particle shape. We chose $\xi = \pm 0.01$, $m = 10$, and $r_0 = 30$ nm. For $\xi = 0.01$, the radius of curvature at the contact point with the substrate is 15 nm, while for ξ

= –0.01, it is 1.48 μm. The scattering of these four particles can be compared to that of a sphere of radius 30 nm. Figure 1 illustrates the five particle shapes. Since the volumes of all of these particles are within 0.12 % of one another, and their function $r(\theta)$ never differs from r_0 by more than 1.2 nm, their free space scattering properties are nearly identical, for both polystyrene and aluminum.

Figure 2 shows the differential scattering cross sections, $d\sigma/d\Omega$ as functions of scattering angle θ_r for the dented spheres, compared to those for a sphere. The polystyrene particles scatter much less than the aluminum particles, and their interactions with their images in the substrate are much weaker. Furthermore, the effect of the sphere deformation is small. If the dent is aimed away from the surface, the scattering function differs from that of the sphere by less than 0.5 %, similar to the changes that occur in free space. If the dent is aimed towards the surface, however, the effect is larger, having about 2.5 % change at large angles, and as much as 6 % change at the scattering minimum near scattering angle $\theta_r = 0°$. Some of what is observed for the polystyrene particles, especially the differential feature near $\theta_r = 0°$, results from the small displacement of the center of the particle towards the surface, and can be predicted by approximations which ignore the near field interaction between the particle and the substrate.

For aluminum particles, the interaction with the substrate is much stronger. While the particle with the dent aimed away from the surface scatters only slightly differently than the sphere (albeit more than that observed for the same shape polystyrene particle), the particle with the dent towards the surface has a significantly different scattering function, different by as much as 36 % from that of the equivolume sphere. These shape dependencies suggest that the shape in the region near the particle-substrate contact point strongly affects the particle scattering function.

Figure 3 shows the results for the Chebyshev particles. Like the dented sphere, the scattering from polystyrene is much less sensitive to the shape of the particle than that from aluminum. The scattering from both aluminum Chebyshev particles differ substantially from that of the spherical aluminum particle, suggesting that the radius of curvature at the contact point, or the contact area, most strongly affects the scattering behavior.

The ratio of the differential scattering cross section of the non-spherical particles to that of the spheres is approximately linear in the contact surface area $A = \pi d(2r_0 - d)$ (for the indented particles) and ξ (for the Chebyshev particles). Thus, for more highly non-spherical particles, the effects shown in Figs. 2 and 3 will be larger. Calculations were also carried out for dented and Chebyshev ellipsoids, and the results were compared to ellipsoids having identical volumes and axis-length ratios. Obtaining similar shape sensitivities as for slightly non-spherical particles, we can conclude that the local shape of a particle near the point where it contacts a substrate will have a profound effect on its scattering properties regardless of the particle's global shape. It is also found that for thinner substrate oxides, the observed effects are even larger than those shown.

A sphere bound to a surface will deform to yield a nonzero contact area. This contact area can be estimated using the theory of Johnson, Kendall, and Roberts (JKR),[11] and depends upon the work of adhesion, the radius of the sphere, and the Young's moduli and Poisson ratios of the materials. For a polystyrene sphere adsorbed on an oxidized silicon substrate, the radius of the contact area can be estimated to be about $a \sim 6$ nm, corresponding to an indentation of $d \sim 0.6$ nm, for a 30 nm radius sphere. The shape of the particle will not exactly match that of the particles described above,[11] but the dented-sphere model provides an estimate of the magnitude of the effects that are expected to occur. The uncertainties in the diameters of polystyrene calibration particles approach 1 % of their diameters,[12] and the differential scattering cross section for a 0.6 nm indentation differs from that of a sphere by about 1 % (half that shown in Fig. 2 for 1.2 nm). Therefore, the finite contact area can have a small but significant impact on the scattering by standard polystyrene spheres, when they are placed on a wafer surface.

The primary concern to those inspecting surfaces for particulate contaminants is the ability for the instruments to correctly size and identify the material. While the scattering from the polystyrene particles is shown to be relatively independent of their shape, as they deviate from spherical, the scattering from the aluminum particles shows a strong dependence on shape. These particles were all chosen to be only slightly different from spherical, and they qualitatively lie well within the range of what most any inspector would want to consider as identical particles. However, the scattering from the aluminum sphere having a dent towards the surface (see Fig. 2) is approximately 20 % to 30 % different than that from the sphere at angles where the scattering is strongest. Such a change in scattering cross section may be incorrectly interpreted by assigning an incorrect size (as much as 5 % error in diameter) or an incorrect material to the detected particle. In conclusion, analysis of light scattering to characterize particle size and material must take into account the possibility that real particles will have non-ideal shapes, which differ only slightly from ideal ones.

REFERENCES

1. P. A. Bobbert and J. Vlieger, Physica **137A,** 209 (1986).
2. T. Wriedt and A. Doicu, Opt. Commun. **152,** 376 (1998).
3. A. Doicu, Yu. Eremin, and T. Wriedt, Opt. Commun. **182,** 281 (2000).
4. P. W. Barber and S. C. Hill, *Light Scattering by Particles: Computational Methods*, (World Scientific, Singapore, 1990).
5. P. Denti, F. Borghese, R. Saija, E. Fucile, and O. I. Sindoni, J. Opt. Soc. Am. A **16,** 167 (1999).
6. J. C. Chao, F. J. Rizzo, I. Elshafiey, Y. J. Liu, L. Upda, and P. A. Martin, J. Opt. Soc. Am. A **13,** 338 (1996).
7. G. Videen, Opt. Commun. **115,** 1 (1995).
8. R. Schmehl, B. M. Nebeker, and E. D. Hirleman, J. Opt. Soc. Am. A **14,** 3026 (1997).
9. Yu. A. Eremin, J. C. Stover, and N. V. Grishina, J. Quant. Spectrosc. Radiat. Transfer. **70,** 421 (2001).
10. Yu. Eremin and N. Orlov, J. Quant. Spectrosc. Radiat. Transfer. **60,** 451 (1998).
11. K. L. Johnson, K. Kendall, and A. D. Roberts, Proc. R. Soc. Lond. A **324,** 301 (1971).
12. G. W. Mulholland, N. P. Bryner, and C. Croarkin, Aerosol Sci. and Technol. **31,** 39 (1999).

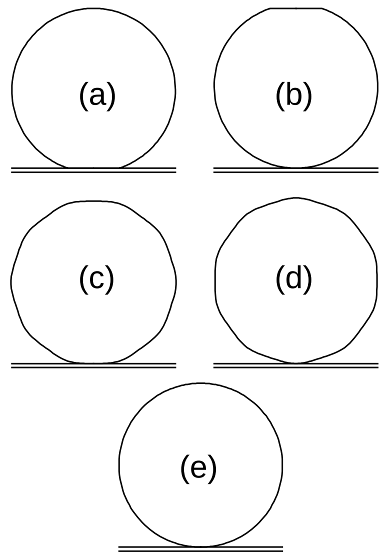

FIG. 1. The profiles of the four particles chosen for this study: (a) 1.2 nm dent towards surface, (b) 1.2 nm dent away from surface, (c) a Chebyshev particle with $\xi = -0.01$, (d) a Chebyshev particle with $\xi = 0.01$, and (e) a 30 nm radius sphere. The 1.5 nm oxide layer on the substrate is shown on the same scale.

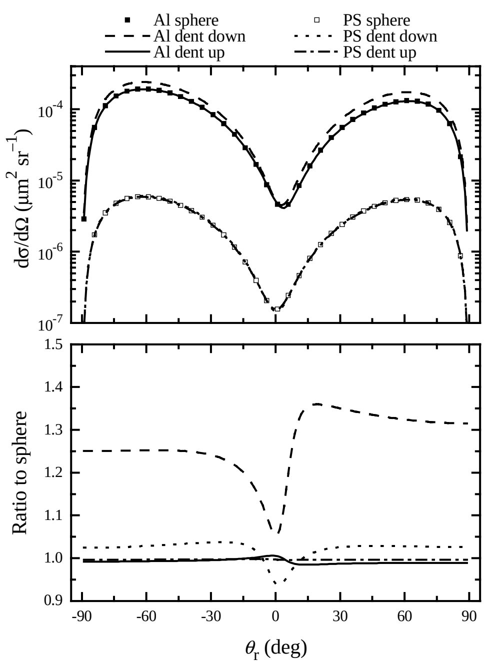

FIG. 2. (top) The differential scattering cross section $d\sigma/d\Omega$ for 30 nm aluminum and polystyrene spheres on a silicon substrate and spheres with 1.2 nm indentations aimed towards the surface and away from the surface. (bottom) The ratios of $d\sigma/d\Omega$ for non-spherical particles to $d\sigma/d\Omega$ for the sphere of the same material. The specular direction is at $\theta_r = 60°$.

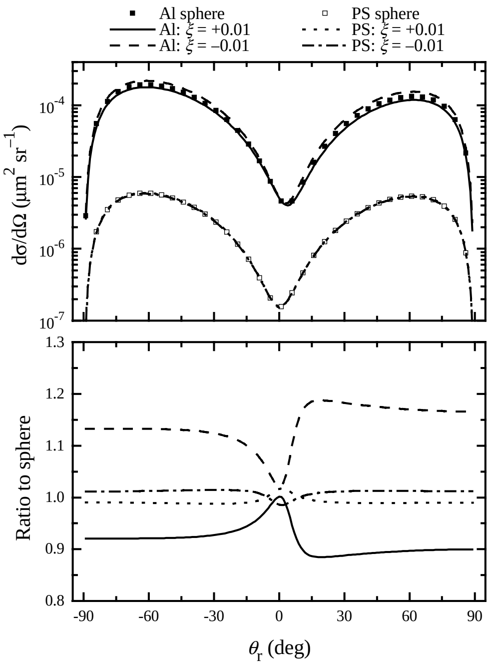

FIG. 3. (top) The differential scattering cross section $d\sigma/d\Omega$ for 30 nm aluminum and polystyrene spheres on a silicon substrate and Chebyshev particles with $\xi = -0.01$ and $\xi = +0.01$. (bottom) The ratios of $d\sigma/d\Omega$ for non-spherical particles to $d\sigma/d\Omega$ for the sphere of the same material. The specular direction is at $\theta_r = 60°$.